SPIN POLARIZED TRANSPORT IN COMPLEX OXIDES

Motivation: A recent EPA estimate finds that office PCs and related electronics are wasting in excess of $2 billion of electricity annually. This wasted energy is responsible for producing as much carbon dioxide as 5 million automobiles[1]. One remedy to this astonishing inefficiency and a means to greater national energy self-sufficiency and a cleaner environment may be found in future electronics platforms based on so-called spintronics devices. In spintronics devices, magnetic spins augment charge in memory and logic elements. To communicate with spins, these nanoscale elements will require the development of 100% spin-polarized injection sources. Today we have identified a class of materials that can provide such fully spin-polarized current—half-metallic colossal magnetoresistive (CMR) manganite oxides. Under the auspices of the CSP program, we propose to explore the basic materials science and physics underlying the magnetic and electronic properties of CMR films and to characterize the critical factors impacting spin polarization in nanoscale devices fabricated from these materials.

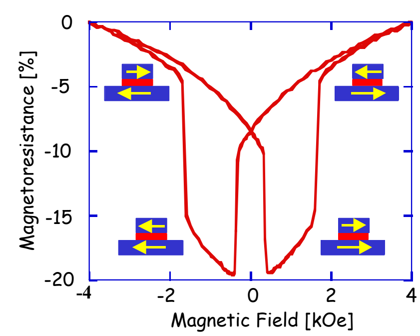

Fig. 1. MR of a $La_{0.7}Sr_{0.3}MnO_3$/oxide/Fe_3O_4 tunnel junction shows values more than an order of magnitude higher than what has been seen in previous epitaxial Fe_3O_4 based junctions. [Hu and Suzuki]

Half-metallic CMR materials only carry current when the spins of the itinerant charges are aligned parallel to the spins of the manganese ions residing in the crystal lattice. This selection rule provides a natural source of ~100% spin-polarized current. The major experimental challenges to fabricating spin polarized devices from these materials relate to:

- interface quality
- tunnel barrier quality
- surface/interface roughness
- ferromagnetic electrode quality magnetic domain walls
- intrinsic behavior of ferromagnetic surfaces/interfaces

Moreover as miniaturization of components continues, a critical challenge to understanding tunnel junction behavior has been to understand the magnetic domain state and magnetization reversal process at the submicron and nanometer length scales. These technological challenges provide a wealth of opportunity for exploring the chemistry and physics underlying these materials- and fabrication-based issues. A focused CSP project combining the experience and capabilities of a consortium of partner institutions is the most efficient way to leverage rapid, comprehensive progress in this high impact field. Our goal will be to address each of the experimental challenges identified above in the context of complex oxide half-metals.

Since submitting this CSP project proposal for consideration in FY01, we have initiated seed projects both at individual institutions and through collaboration among our team members. In addition to identifying the vision and scope of our CSP project, this preproposal will highlight examples of these new connections.

Materials: Materials synthesis and processing issues are critical to success in this endeavor. For example, transport behavior in CMR device structures has been plagued with issues of inhomogeneity.[2-5] Do these inhomogeneities arise intrinsically (e.g., intrinsic phase separation), through interface scattering, or through some other mechanism entirely? Patterned structures geometrically confined to submicron length scales will shed light on the half metallicity and magnetotransport as a function of the strain state and how it is affected by phase separation or other intrinsic inhomogeneities. Our experience in bulk naturally layered manganites (ANL) will facilitate selection of materials designed to address this and related issues. While much thin film growth is based on the ability to isolate a target stoichiometry in bulk form, thin film deposition may result in the synthesis of metastable phses with unexpected properties that may not be observed in bulk form. For example, we can identify compositions which are expected to exhibit desirable materials properties but which may be thermodynamically inaccessible to bulk crystal growth. The kinetic control of film synthesis may allow access to these desirable metastable products. The search for new half-metallic ferromagnets will be an important aspect of this project.

Nanostructures: Our goal is to investigate the basic synthetic techniques, fabrication science, and magnetoelectronic properties of CMR-based nanostructures. We will study nanostructures where lateral confinement, combined with layering in the form of naturally layered compounds and/or artificial multilayers, will give rise to a wide range of magnetic and electronic behavior. The nanostructures will be patterned lithographically from films deposited by a variety of techniques, including pulsed laser deposition (PLD), sputtering, laser molecular beam epitaxy (MBE), and atomic layer-by-layer deposition. Close collaboration among the participating institutions is critical, as each brings its own expertise in particular synthesis techniques. In addition, we will build on a materials knowledge-base growing out of bulk layered materials (e.g., naturally layered manganites) for both materials choice and precursor targets. Our experience with this class of materials will position us uniquely among spintronics programs worldwide.

Fabrication of patterned samples will draw upon existing and soon to be available technology in Centers for Nanoscience sponsored by the DOE and at partner universities. Future advances in electron beam lithography will allow us to fabricate junctions reproducibly with lateral dimensions of 100 nm. In addition, at ANL we propose to use the atomic force microscope (AFM) as a novel processing tool to fabricate new materials at the nanoscale. Moving the AFM tip across a surface with a controllable force will selectively remove material allowing us to achieve resolution down to 20 nm.

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Scanning probe techniques will be used to determine the morphology of spintronic architectures with unit cell height resolution and their magnetic properties. Local STM spectroscopies (ORNL, UIUC) will complement both photoemission (BNL) and bulk magnetotransport measurements on thin film samples and prototype nanostructures.

Projects: Our experience in bulk naturally layered manganites suggests a number of potential projects designed to exploit the strong composition dependence of charge, lattice, and magnetic states. As a world-leader in the synthesis and crystal growth of these materials, we bring a unique focus to spintronics research not found in competing programs. Examples of future directions for the project include:

- *Spin-valve effects*. Current generation spintronics are based on the GMR effect and its associated spin-valve structures of metallic multilayers. Certain compositions of layered manganites exhibit an *intrinsic* spin-valve effect. Studies in single crystals (ANL) and etched mesas on single crystals [7] demonstrate these effects. Initially these crystals can provide a platform to conduct fundamental studies using layered structures that are virtually perfect at the atomic scale.
- *Hybrid materials*: The ground state of doped CMR oxides is extremely sensitive to the dopant concentration. In addition, due to strong spin/charge/lattice interactions, strain coupling provides the possibility of active control of magnetic and/or charge ordering. As a Center research focus, we propose to take active control of strain using patterned or self-assembled arrays of nanocomposites comprised of a ferroelectric host and a CMR gues. Through appropriate materials choices, we will investigate (a) continuous variation of strain coupling *via* applied field, (b) uniaxial strain control to modify the anisotropy of orbital ordering and its associated magnetoelectronic response, and (c) relative phase stability by exploiting the delicate balance between bulk and surface/interfacial energies. These approaches are a developing part of the long-term strategy at both ANL and ORNL.

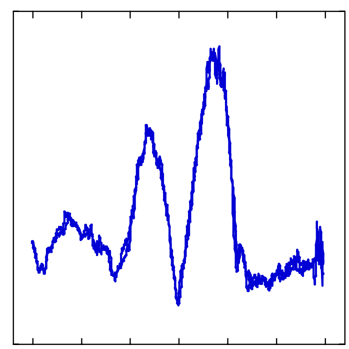

Fig 2. Point contact tunneling measurement of naturally layered manganite crystal measured at ANL.

Center Participants and Strategy: Principal investigators at six national labs (ANL, BNL, LANL, LBL, LLNL, ORNL) and two universities (UIUC, UT) make up our team. A list of participants is attached.

Studies of materials preparation will be broadly pursued throughout the Center, while technique-specific experiments will be assigned to institutions with expertise and infrastructure.

- Deposition of thin film manganites will be pursued through several parallel programs, including PLD efforts at Berkeley (Y. Suzuki), ANL (Miller), LANL (Q. Jia), laser MBE techniques at ORNL (J. Shen), sputtering at ANL (S. Bader), and atomic layer-by-layer growth at Illinois (J. Eckstein). Specific research directions for each of these efforts will be assessed throughout the project lifetime to avoid duplication and to allow access to a broad range of materials systems.
- Expertise in transport measurements of anisotropic single crystals at ANL will enable the development and study of mesa structures using true nanoscale patterning that pushes beyond the projected resolution of resist-based lithography. These studies will be paralleled with microsopic transport measurements on CMR films at ORNL (A. Baddorf, J. Wendelken).
- Scanning probe experiments at ORNL will explore the local electronic structure and its relationship to inhomogeneities, frustration, etc., while BNL expertise in spin-polarized photoemission will enable exploration of the magnetic/electronic state of our structures.
- In addition to synthesis, patterning, and properties assessment, a strong theory component will be a critical aspect of this Center. Here expertise in the theory of spin injection (D. Smith, LANL) will join with strong in-house theory at ORNL and ANL to understand growth modes and competing ground state structures.

Highlights of Seed Projects: During the past months since our application for CSP funding in FY01, several participants have begun seed projects both individually and collaboratively. Examples include scanning probe studies of naturally layered manganite crystal surfaces (ANL/UIUC, Fig. 2) and magnetic depth profiling via neutron reflectometry on exchange bias manganite superlattices (LANL/ANL). These seed projects reflect a commitment on the part of the project participants to the proposed science as well as to forging collaborative links.

Technology Partnering: ANL groups working in ferroelectric materials (Streiffer) are well connected to DoE technologies-funded programs. Furthermore, discussions with potential industrial resources (J.Z. Sun, IBM; John Slaughter, Motorola) have led to their support of this program. The exact role of such partners will be defined as the full proposal is developed. It is noteworthy that many of the canonical CMR materials, such as $La_{1-x}Sr_xMnO_3$, are also of interest to DOE-supported technologies outside the purview of spintronics, including solid oxide fuel cells. Existing applied programs at several of the national labs may become involved both as consultants and as "spin-off" beneficiaries of our materials research.

References

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